Multi-model assessment of the factors driving stratospheric ozone evolution over the 21st century


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Abstract

The evolution of stratospheric ozone from 1960 to 2100 is examined in simulations from fourteen chemistry-climate models. There is general agreement among the models at the broadest levels, showing column ozone decreasing at all latitudes from 1960 to around 2000, then increasing at all latitudes over the first half of the 21st century, and latitudinal variations in the rate of increase and date of return to historical values. In the second half of the century, ozone is projected to continue increasing, level off or even decrease depending on the latitude, resulting in variable dates of return to historical values at latitudes where column ozone has declined below those levels. Separation into partial column above and below 20 hPa reveals that these latitudinal differences are almost completely due to differences in the lower stratosphere. At all latitudes, upper stratospheric ozone increases throughout the 21st century and returns to 1960 levels before the end of the century, although there is a spread among the models in dates that ozone returns to historical values. Using multiple linear regression, we find decreasing halogens and increasing greenhouse gases contribute almost equally to increases in the upper stratospheric ozone. In the tropical lower stratosphere an increase in tropical upwelling causes a steady decrease in ozone through the 21st century, and total column ozone does not return to 1960 levels in all models. In contrast, lower stratospheric and total column ozone in middle and high latitudes increases during the 21st century and returns to 1960 levels.
1. Introduction

The evolution of ozone in the 21st century is a critical science issue. While changes in ozone are presently controlled primarily by declines in halogen concentrations, variations in temperature, circulation, and oxides of nitrogen and hydrogen also affect ozone [WMO 2003; WMO 2007]. Throughout the stratosphere there are long-term changes in various processes and the balances among them. Therefore it is difficult to find a single approach to identify the contributions of different mechanisms affecting ozone levels throughout the stratosphere.

Ozone loss throughout much of the stratosphere was dominated by rising halogen concentrations until about 1997, the time of peak halogen loading [e.g., Yang et al., 2006; Shepherd and Jonsson, 2008; Yang et al., 2008]. In the 21st century, as halogen concentrations are expected to decrease at about 1/3 of the rate of their increase in the late 20th century, other factors will likely play a more significant role in the evolution of ozone. Stratospheric cooling will decrease the rate of gas-phase reactions that destroy ozone, and thereby increase concentrations of ozone [e.g., Haigh and Pyle, 1979; Brasseur and Hitchman, 1988; Shindell et al., 1998; Rosenfield et al., 2002]. Future increases in N2O and CH4 increase loss of ozone by nitrogen and hydrogen catalytic cycles, [e.g., Randeniya et al., 2002; Rosenfield et al., 2002; Chipperfield and Feng, 2003; Portmann and Solomon, 2007; Ravishankara et al., 2009] but in a changing climate, increases in N2O do not necessarily cause NOy (or NOx) to increase proportionately throughout the stratosphere. Rosenfield and Douglass (1998) showed that NOy loss rates increase as the stratosphere cools, which will impact future levels of ozone. Several studies have linked increases in greenhouse gases (GHGs) to changes in stratospheric transport,
further impacting ozone recovery [Waugh et al., 2009; Li et al., 2009; Hegglin and Shepherd, 2009].

Eyring et al. [2005] laid the groundwork for the Chemistry-Climate Model Validation (CCMVal) activity in which numerous chemistry-climate models (CCMs) were evaluated to increase our confidence in predicting future stratospheric ozone change. The results of the CCMVal-1 simulations of past ozone changes conducted for this activity were presented in Eyring et al. [2006], which evaluated processes important in determining the distribution of ozone. In a follow on study Eyring et al. [2007] presented the projections of stratospheric ozone change over the 21st century simulated by these CCMs, and discussed how quantities that can impact ozone are projected to change.

CCMVal-1 was the first major coordinated activity where an ensemble of CCMs performed simulations with similar external forcings to assess ozone evolution [Eyring et al., 2007]. The CCMVal-2 activity [Eyring et al., 2008] included more models (14 are used here) and, in addition, more simulations cover the entire period of interest (1960-2100) than in CCMVal-1 (3 models). Here, we make use of CCMs that participated in the CCMVal-2 activity and that contributed projections of stratospheric ozone evolution until the end of the 21st century. These simulations are based on observed GHG and halogen concentrations in the past and on one predicted scenario for the future.

Austin et al. [2010a] examined the evolution of total column ozone and compared ozone and Cl, recovery dates in the CCMVal-2 models. Here we (a) contrast the evolution in the upper and lower stratosphere, and (b) examine the causes of the ozone changes (and differences among the models). To accomplish this, we apply a variety of methods depending on the physical and/or chemical processes that dominate ozone evolution in various regions. Section 2 gives an
overview of the models and model simulations. The results are discussed for extra-polar ozone in Section 3 and polar ozone in Section 4. Variations in ozone recovery by region are discussed in Section 5 and a summary of the conclusions is given in Section 6.

2.1 Models and Model Simulations

CCMVal-2 model simulations were conducted to improve the understanding of models through process-oriented evaluation along with discussion and coordinated analysis [SPARC, 2010]. Information about individual model simulations and references for each are summarized in Table 1. In addition, Morgenstern et al. [2010] present a much more detailed overview of the models that participated in this activity. Here we consider 1960 to 2100 simulations from the CCMs whose domains include at least the upper stratosphere, and can then be used to contrast the ozone evolution in the lower and upper stratosphere. All models in this study have output for 1960-2099, except UMUKCA-METO which ends in 2083.

REF-B2 uses the A1b GHG scenario from IPCC [2000] and the adjusted A1 halogen scenario from WMO [2007]. The adjusted A1 halogen scenario includes the earlier phase out of hydrochlorofluorocarbons mandated by recent revisions to the Montreal Protocol. For GEOSCCM a combination of REF-B1 (1960-2000) and REF-B2 (2001-2099) was used. REF-B1 differs only in that observed Hadley sea surface temperatures (SST) and sea ice data [Rayner et al., 2003] were used instead of modeled SSTs. All model runs with the exception of CMAM use prescribed sea surface temperatures and sea ice extent based on fully coupled atmosphere-ocean runs for the A1b scenario. CMAM includes a fully coupled ocean model in its
All models use sophisticated stratospheric chemistry schemes except AMTRAC3, in which halogens are not explicitly modeled but rather Cl$_y$ and Br$_y$ are parameterized [Austin and Wilson, 2010; Morgenstern et al., 2010]. REF-B2 simulations use background non-volcanic aerosol loading and there are no imposed solar cycle variations. Although several modeling groups submitted multiple ensemble simulations, only one member from each group was used in this analysis because the intermodel differences are generally much larger than the ensemble spreads [Eyring et al., 2007]. The model output used in this study is a zonal mean monthly mean average. The partial and total column ozone amounts are integrated over the standard CCMVal-2 pressure levels.

In order to view all 14 models on a single plot, filtering is necessary to remove short-term (e.g. high frequency) variations. In all cases (except where noted) a 1:2:1 filter is used iteratively 30 times as described in Eyring et al. [2007] to smooth the model output displayed in the figures. While some differences can be seen in individual models between this filter and the time series additive-model (TSAM) analysis, they are typically very small and do not impact our conclusions [Scinocca et al. 2010, in preparation].

2.2 MLR Analysis

One of the primary methods we use to estimate the contribution of different mechanisms to the simulated changes in ozone is multiple linear regression (MLR). The method is explained in detail in Oman et al. [2010], but we repeat some of the basic details here. For a given location and time, MLR is applied to determine the coefficients $m_x$ such that

$$\Delta O_3(t) = \sum_j m_x \Delta X_j(t) + \varepsilon(t),$$  

(1)
where the $X_j$ are the different quantities that could influence ozone, the coefficients $m_x$ are the sensitivity of ozone to the quantities $X$, i.e., $m_x = \partial O_j / \partial X$, and $\varepsilon$ is the error in the fit. To do this four explanatory variables ($X_j$) are used in (1): (i) Cl$_y$ + α Br$_y$, (ii) reactive nitrogen (NO$_y$

\[
= \text{NO + NO}_2 + \text{NO}_3 + 2*\text{(N}_2\text{O}_5) + \text{HNO}_3 + \text{HO}_2\text{NO}_2 + \text{ClONO}_2 + \text{BrONO}_2, \]

(iii) reactive hydrogen (HO$_x$

\[
= \text{OH + HO}_2), \text{ and (iv) temperature (T). Each term on the right hand side of equation (1) gives the “contribution” of the response in ozone due to a change in $X$. We use $\alpha = 5$ in the definition as Daniel et al. [1999] show this is an appropriate value for the upper stratosphere, which is the chemically driven region for which the MLR analysis is most appropriate. A test of this method using $\alpha = 60$ did not noticeably impact any of the individual contributions calculated in this study. We do use $\alpha = 60$ in section 4 when examining polar lower stratospheric ozone changes.

There are several limitations with the above linear regression approach as discussed in Oman et al. [2010] which are summarized below. First, other mechanisms that are not considered in the regression (e.g., transport) could play a role. Second, significant correlations can exist between the temporal variations of the quantities, i.e., the quantities are not necessarily independent. Third, a high correlation between ozone and a quantity does not show causality, as ozone could be causing the quantity to change, or changes in another quantity could be causing both ozone and the quantity of interest to change in a correlated way. Temperature and ozone in the upper stratosphere are an example of this third complication: changes in ozone cause changes in temperature through changes in short-wave heating [Shepherd and Jonsson, 2008]. At the same time, the local ozone concentration responds to changes in temperature by changing chemical reaction rates. Also, the relationship between the regression variables and ozone may not be linear. For example, the ClO/Cl$_y$ ratio varies as a function of temperature in a highly non-linear manner. One could use a regression based on the rates of odd-oxygen loss by nitrogen,
chlorine, and bromine radical abundances as regressor variables, as was done by Yang et al., [2006]. However, this treatment is beyond the scope of the present analysis, as the information archived by each model makes use of radical abundances challenging. Because of the above limitations, caution must be applied when interpreting the MLR results presented below.

UMUKCA-UCAM and AMTRAC3 did not provide HO$_x$ so for these models the MLR consists of three explanatory variables. This analysis has been repeated without HO$_x$ for all models and while there is some decrease in explained variance from the MLR the main findings and conclusions are not impacted.

### 3. Extra-Polar Ozone

#### 3.1 Total column ozone

Austin et al. [2010a] examined the changes in total column ozone in the CCMVal-2 simulations. Here, we start with total column ozone and then break down the ozone amounts into two partial columns for the extra-polar region. Figure 1a-c shows the evolution of total column ozone amounts over the tropics (25°S-25°N) and midlatitudes of each hemisphere (35-60°S and 35-60°N), for each of the CCMVal-2 models. The evolution is shown with respect to 1960 levels, and has been smoothed as described above. Also shown are ground-based total column ozone measurements (solid black curves, updated from Fioletov et al., [2002]) with respect to the smoothed 1964 value.

There is qualitative agreement in the evolution of ozone between regions and models at the broadest level, with ozone decreasing from 1960 to around 2000, and then increasing in the
first half of the 21st century [Austin et al., 2010a]. All models consistently indicate tropical total
column ozone at the end of the 21st century is less than it was in 1960 (Figure 1b). In contrast,
total column ozone over the midlatitudes (Figures 1a,c) in 2100 exceeds 1960 values in nearly all
the models, with a generally larger increase in Northern Hemisphere (NH) mid-latitudes.

There is a large spread in the magnitude of the peak ozone loss (around 2000) and the
date that column ozone is predicted to return to 1960 levels. For example, in SH mid-latitudes
the simulated decrease in ozone from 1960 to 2000 varies from around 10 DU to over 50 DU,
while the date of return to 1960 levels for the same region varies from around 2030 to after 2100.
In all three latitude bands CNRM-ACM and MRI show the largest loss in total column ozone by
the year 2000, and also tend to have later return dates. UMUKCA-METO has the smallest ozone
loss in the tropics and Southern Hemisphere (SH), and one of the smallest decreases in NH
mid-latitudes. The reasons for these model differences are examined in Section 3.2. Also, there
is a large difference in the ozone decline in the SH midlatitudes prior to 1975 found by the
various models (declines ranging from 0 to 20 DU), with no change over this time period
apparent in the ground-based ozone observations. Ground-based total column ozone observations
indicate about a 20 DU decrease by 2000 in the SH midlatitudes and about a 15 DU decrease in
the NH midlatitudes. In the tropics only small total column ozone changes have been observed
and are less than the model-simulated changes with the exception of UMUKCA-METO, which
are similar.

3.2 Partial column ozone
While the evolution of total column ozone and its impact on surface UV radiation changes are of great importance, it is useful to look at changes in the partial columns [e.g., Yang et al., 2006; Li et al., 2009]. This shows the relative role of changes in the upper and lower portions of the stratosphere in the differences described above. As in Oman et al. [2010] we split the stratosphere at 20 hPa because the ozone changes at 20 hPa are generally small, and 20 hPa separates the photochemically controlled ozone region above from the transport and chemically driven region below. Other studies have used 15 hPa [e.g. Li et al., 2009] or 25 km altitude (~25 hPa) [e.g., Yang et al., 2006] but we prefer 20 hPa, as used in Oman et al., [2010].

Figures 1d,e,f and 1g,h,i show the evolution of the partial column ozone amounts for the upper (20 to 0.1 hPa) and lower (500 to 20 hPa) portions of the column. The evolution in the upper partial column ozone is similar among the 3 regions, but there are significant differences in the lower partial column. It is the differences in the lower stratosphere that cause differences in the evolution of column ozone between regions. In particular, the decrease in tropical column ozone over the latter half of the 21st century is due to decreases in the lower stratosphere (Figure 1h), which are larger than increases in upper stratospheric ozone (Figure 1e).

There are quantitative differences in the upper columns but these are generally smaller than the differences in the lower columns, especially in midlatitudes (note the different scales for upper and lower column plots in Figure 1). Peak losses occur around the year 2000, averaging 3 to 7 DU over tropical and midlatitudes for the upper partial column of most models, with MRI and CNRM-ACM having losses around 10 DU. In the lower partial column there are much greater differences in the ozone change over this range of latitude bands. Midlatitude ozone losses around 2000 are largest in the SH, as expected due to the effects of the Antarctic ozone hole [Atkinson et al., 1989]. Ozone losses range from 10-25 DU with less ozone loss in
UMUKCA-METO and significantly more in MRI and CNRM-ACM. Tropical latitudes show
largest loss of ozone by the end of the 21st century, with a range of 7 to 18 DU. The tropical
lower partial column is the only region where ozone is consistently forecast to continue to
decline until the end of the 21st century, due to increased strength of the Brewer-Dobson
circulation in the models that is driven by rising GHG concentrations [Rind et al., 1998; Buchart
and Scaife, 2001; Butchart et al., 2006; Shepherd, 2008; Li et al., 2009]. It should be noted that
models with larger (smaller) total column ozone changes generally have larger (smaller) changes
in both lower and upper partial columns.

3.3 Upper Stratosphere

As discussed above, and shown in Figure 1d-f, the evolution of ozone in the upper portion of the
stratosphere is qualitatively similar among most models. Also, the overall behavior of ozone in
the tropics and midlatitudes (both hemisphere) is similar. Nonetheless, there are quantitative
differences among the models. We use the MLR analysis described in Section 2 to examine the
mechanisms causing the ozone changes, and the differences among the models.

We consider first the evolution of ozone at 5 hPa in the tropics (25°S-25°N). Figure 2a
shows the evolution of the ozone mixing ratio while Figure 2b shows the change in ozone with
respect to 1960 levels. There is a large spread in the time-mean values of ozone among the
models (from around 8 ppmv to around 11 ppmv), and also some differences in changes relative
to 1960 levels. All models show a peak ozone loss around 2000, with magnitudes of 0.5 and 0.7
ppm for most models. The MRI and CNRM-ACM models show the largest peak ozone loss
(greater than 1.0 ppm), whereas AMTRAC3 and LMDZrepro have the smallest loss (less than
0.4 ppm). All models also show 5 hPa ozone returning to 1960 levels before the end of the 21st century, but there is a large spread in dates when this occurs (from early 2020s to 2060s).

As presented in the Introduction, several mechanisms can cause changes in ozone, including changes in halogens, temperature, reactive nitrogen and hydrogen, and transport. Figures 2c,e,g,i show the evolution of Cl\textsubscript{y}+\textalpha Br\textsubscript{y}, temperature, NO\textsubscript{y}, and HO\textsubscript{y}, respectively. The evolution of Cl\textsubscript{y}+\textalpha Br\textsubscript{y} and variation among models (Figure 2c) is somewhat similar to that of ozone (Figure 2a), suggesting that differences in Cl\textsubscript{y}+\textalpha Br\textsubscript{y} can explain much of the ozone evolution. However, there is not a simple one-to-one relationship between changes in ozone and those in Cl\textsubscript{y}+\textalpha Br\textsubscript{y}. For instance, ozone exceeds 1960s values in the later part of the 21st century, while Cl\textsubscript{y}+\textalpha Br\textsubscript{y} is still above 1960 levels. Also, models with larger values of Cl\textsubscript{y}+\textalpha Br\textsubscript{y} (Figure 2c) do not necessarily have larger ozone loss, e.g., MRI and CNRM-ACM have the largest peaks in ozone loss, but do not have the largest changes in Cl\textsubscript{y}+\textalpha Br\textsubscript{y}.

One possible cause for the lack of a simple relationship between ozone and halogen changes is differences in temperature trends. As shown in Figure 2e, all models show cooling throughout the 21st century, largely from increasing CO\textsubscript{2} concentrations, and this is expected to increase ozone (by slowing down the rate of reactions that destroy ozone). However, it should be noted that over the recent past about 50% of the cooling is associated with the ozone loss from Cl\textsubscript{y}+\textalpha Br\textsubscript{y} [Shepherd and Jonsson, 2008]. This cooling explains the increase in ozone above 1960 levels before Cl\textsubscript{y}+\textalpha Br\textsubscript{y} returns to 1960 values. Most models cool by about 8-9 K below 1960 levels by the end of century, but a few models have cooling exceeding 10 K (CCSRNIES, UMSLIMCAT, LMDZrepro, and CMAM). This is consistent with an expected increase in ozone due to cooling, and these four models have among the largest ozone increases by 2100, as shown in Figure 2b.
To quantify the relative contributions of the above mechanisms to ozone changes we now apply an MLR analysis to model outputs. As described in Section 2, the MLR analysis is used to quantify the contributions of changes in Cl\textsubscript{y}+\alpha Br\textsubscript{y}, temperature, NO\textsubscript{y}, and HO\textsubscript{x} to the ozone changes. As previously discussed in Section 2, there are a number of caveats with this type of analysis and care should be taken in interpreting the results.

The contributions from Cl\textsubscript{y}+\alpha Br\textsubscript{y}, temperature, NO\textsubscript{y}, and HO\textsubscript{x}, are shown in Figures 2d,f,h,j, respectively. Differences among models in the contribution from Cl\textsubscript{y}+\alpha Br\textsubscript{y} are generally similar to relative variations in Cl\textsubscript{y}+\alpha Br\textsubscript{y} (compare Figures 2d and 2c), with some exceptions. Most notably, MRI and CNRM-ACM have the largest ozone loss at this level from 1960-2000 despite not having unusually large amounts of Cl\textsubscript{y}+\alpha Br\textsubscript{y}. This is related to larger sensitivities to Cl\textsubscript{y}+\alpha Br\textsubscript{y}, and we quantify this sensitivity below.

Temperature trends play an important role in the return of ozone to above historical levels: the four models with the largest cooling trend (LMDZrepro, CCSRNIES, CMAM, and UMSLIMCAT, see Figure 2e) have the largest increases in ozone relative to 1960 levels (see Figure 2b). The differences in cooling among models explain most of the differences in projected ozone by 2100. Although, again, we note that it is not a simple one-to-one relationship.

The contributions from NO\textsubscript{y} and HO\textsubscript{x} changes are significantly smaller in most cases. Figure 2h,j show the contributions of NO\textsubscript{y} and HO\textsubscript{x} to ozone changes. For NO\textsubscript{y} most models show losses of 0.2-0.3 ppm by 2100 with a few models showing somewhat larger loss, up to a loss of 0.7 ppm in AMTRAC3. While 5 hPa is typically one of the largest loss regions from NO\textsubscript{y} changes in volume mixing ratio, HO\textsubscript{x} has a much smaller impact at this level and typically is more important at altitudes above 5 hPa [Jackman et al., 1986]. All models show a relatively
small contribution (less than 0.1 ppm) from HO, except ULAQ, which has the largest HO, trend (see Figure 2i). In general, for the A1b greenhouse gas scenario considered in these CCMVal-2 simulations, NO and HO, contributions are much smaller than the contributions from halogen recovery and stratospheric cooling over the 21st century. It is however important to note that this may not be the case for other GHG scenarios [e.g., Oman et al., 2010].

The above analysis indicates that changes in Cl,+Br, and temperature dominate tropical ozone evolution at 5 hPa, and differences in the simulated changes in these quantities explain most (but not all) of the variations among the models. To see how representative the changes at 5 hPa are over a range of pressures, Figure 3 shows vertical profiles of changes between 2000 and 2100 for 25°S-25°N.

Figure 3a shows each models reference ozone profile for the year 2000 while changes by 2100 appear in Figure 3b. Also, plotted on Figure 3a are Microwave Limb Sounder (MLS) satellite measurements (solid black curve) averaged from 2005-2009. The peak ozone increase is typically at 3 hPa (Figure 3b) with most models centered at around 1.5 ppm. Figure 3c shows that in the upper stratosphere most models have a Cl,+Br, decrease of 2 ppb over this time period. CCSRNIES and SOCOL have larger than average decreases in Cl,+Br, and AMTRAC3 has a smaller than average Cl,+Br, change with a different vertical structure, which is due to how AMTRAC3 treats the breakdown of halogens. Again, we see that MRI and CNRM-ACM have the largest ozone changes due to Cl,+Br, (Figure 3d) with AMTRAC3 having the smallest increase over the upper stratosphere. Most models show about equal contributions from decreases in Cl,+Br, and decreases in temperature to the ozone increases over the 21st century. Observations from the MLS [Santee et al., 2008a] for HCl (not shown)
suggest that while a few models are in good agreement with MLS in the upper stratosphere, many appear to be on the low end with only CCSR-NIES on the high end of observations.

Figure 3 shows that the key features seen at 5 hPa hold throughout the upper stratosphere (10 – 1 hPa). Models that have high (low) ozone changes at 5hPa have similar characteristics throughout the upper stratosphere. Also, differences in Cl$_y$+αBr$_y$ can explain most of the variations among the models with additional contributions from temperature. Figure 3h shows the contribution from NO$_y$ to changes in ozone: it is typically much smaller, and opposite in sign, than changes due to Cl$_y$+αBr$_y$ or temperature. In general, models show NO$_y$-related ozone loss of 0.2 ppm around 3-10 hPa, with only AMTRAC3 exhibiting a larger loss. As presented in the Introduction, stratospheric cooling increases NO$_y$ loss [Rosenfield and Douglass, 1998] causing a smaller increase (and even some decreases, see Figure 3g) in NO$_y$ over the 21st century than would be expected from N$_2$O increases in the absence of climate change. The interpretation of changes in lower stratospheric ozone are complicated by the increasingly important impact of transport which is not explicitly included in the MLR analysis and will be discussed in more detail in Section 3.4.

As discussed above there are not always one-to-one relationships between changes in Cl$_y$+αBr$_y$ and temperature and their contribution to ozone changes. One possible reason for this is the different sensitivities of ozone to Cl$_y$+αBr$_y$ and temperature. Figure 4 shows the vertical profiles of the sensitivity of tropical ozone (25°S-25°N) to Cl$_y$+αBr$_y$, temperature, and NO$_y$ calculated over the period 1960-2100. Overall there is good agreement between the sensitivities of the various models. All models show a peak Cl$_y$+αBr$_y$ sensitivity at 3 hPa, mostly around -0.35 ppm/ppb. However, two models stand out with much larger sensitivities, CNRM-ACM (-0.6 ppm/ppb) and MRI (-0.8 ppm/ppb) at 3 hPa.
Further insight into the sensitivity of modeled ozone to Cl$_y$+Br$_y$ can be gained by examining the abundance of ClO and BrO archived by the various CCMs. Here, we provide a snapshot of the analysis presented in Chapter 6 of SPARC CCMVal [2010]. Figure 5 shows profiles of the ClO/Cl$_y$ ratio archived by four of the CCMs at 35°N for Sept 1993 and at 22°N for Feb 1996 (colored lines). The black lines represent the calculated ratio of ClO/Cl$_y$ found using a photochemical steady state (PSS) box model (e.g., Canty et al., 2006 and references therein) constrained by fields of O$_3$, H$_2$O, CH$_4$, Cl$_y$, NO$_y$, aerosol surface area, and other constituents archived by each CCM group. Results are shown for four models: the two in question (MRI and CNRM-ACM) and two that archive ClO values that are well explained by the PSS simulation (CMAM and WACCM). The numerical value on each panel is the “metric” assigned to each comparison, with a value of unity indicating perfect agreement with the PSS simulation.

The profiles of ClO archived by CMAM and WACCM are in excellent agreement with the PSS simulation, indicating that implementation of the chemical mechanism within these models agrees well with the implementation of the mechanism within the PSS model. However, the MRI ClO profile is much higher (green dashed line, Figure 5) than the profile found using the PSS model. As explained in SPARC CCMVal [2010], the MRI chemical mechanism neglected to include the channel for the loss of ClO by the reaction of ClO+OH to yield HCl+O$_2$. This leads to much higher values of ClO than are reported by other models, or observed in the atmosphere [Santee et al., 2008b]. Even when we neglect this reaction channel in our PSS model (black dotted curves, Figure 5), we still can not account for the high values of ClO archived by the MRI group. We conclude the high sensitivity of O$_3$ to Cl$_y$+Br$_y$ exhibited by the MRI model is due to the high concentrations of ClO in this model, due in part to the neglect of the ClO+OH$\rightarrow$HCl+O$_2$ channel.
The reason for the high sensitivity of $O_3$ to $Cl_y + \alpha Br_y$ within the CNRM-ACM model is not known. For most altitudes, $ClO$ within CNRM-ACM is simulated well by the PSS model. CNRM-ACM does simulate much higher values of $ClO$ just above the tropopause than are accounted for by the PSS simulation; within CNRM-ACM, it appears that chlorine activation is occurring for warmer conditions than suggested by the PSS comparison. The high $ClO$ just above the tropopause cannot explain the large sensitivity of $O_3$ within this model to $Cl_y + \alpha Br_y$ shown in Figures 2d, 3d, and 4a. We have examined profiles of $BrO$ within the CNRM-ACM and the PSS model (not shown); we do not believe, based on this comparison, that the high sensitivity of $O_3$ to halogens exhibited by CNRM-ACM is due to the calculation of $BrO$ within this CCM. It is important to note that the PSS comparisons were done for the REF-B1 simulation (1960 to 2000, observed aerosols) whereas the bulk of this analysis is based on the REF-B2 simulation (1960 to 2100, background aerosols). Finally, our comparisons cannot reveal possible errors in the coding of a rate constant that would affect the rate of ozone loss by $ClO$ or $BrO$, but not affect the abundance of $ClO$ or $BrO$ (i.e., the reaction of $ClO+O$). The reason for high sensitivity of $O_3$ to $Cl_y + \alpha Br_y$ within the CNRM-ACM model remains unexplained.

We performed an uncertainty analysis of the sensitivities as in Oman et al. [2010] and find similar results (not shown). That is, in the mid to upper stratosphere there is relatively more uncertainty in $NO_y$ sensitivity compared to much smaller uncertainty associated with $Cl_y + \alpha Br_y$ and temperature.

The evolution of upper stratospheric ozone and of the different contributing ozone loss mechanisms at midlatitudes is very similar to that in the tropics (not shown). There are again, reasonably balanced, equal contributions of $Cl_y + \alpha Br_y$ and temperature for the midlatitudes as we
found in the tropics. Again MRI and CNRM-ACM stand out with much larger contributions from Cl$_2$+Br$_2$ to ozone changes.

3.4 Lower Stratosphere

As shown in Figure 1 the evolution of ozone in the tropical lower stratosphere (p > 20 hPa) differs from that in the upper portion. This can also be seen in the profiles shown in Figure 3b.

Although an MLR analysis has been applied to the lower stratosphere, interpretation of these results is complicated by the large role of transport changes and the tight coupling of upwelling and temperature. In the tropical lower stratosphere the loss that the MLR attributes to a temperature change is actually largely a response to increased upwelling, which acts to decrease ozone. This is consistent with Avallone and Prather [1996], who showed that changes in upwelling are dominant in controlling tropical lower stratospheric ozone. Temperatures can be reduced by both increased in upwelling and decreased ozone. Previous studies have shown that an increase in the tropical upwelling is the principal cause of a decrease in tropical ozone [Shepherd, 2008; Li et al., 2009] with an additional possibly smaller contribution from “reverse self-healing”. Reverse self-healing occurs as upper stratospheric ozone rises above historical levels causing less ultra-violet radiation to penetrate into the lower stratosphere resulting in the decreased production of ozone.

We examine here the relationship between tropical upwelling and ozone in the CCMVal-2 models. Figure 6 compares the change in tropical ozone (25°S-25°N) at 50 hPa between 1960 and 2100 to the corresponding change in the vertical residual velocity ($w^*$) at 70 hpa, for the 12
models that provided \( w^* \) fields. All models show an increase in tropical upwelling and a
decrease in ozone over this time period. However, there is a large spread amongst the models in
the change in both quantities. There is a fairly compact relationship between these two quantities
and a linear fit (black line) nearly intersects the origin indicating that increases in upwelling are
the dominant contributor to ozone loss at this level. Most models indicate increases in upwelling
of 0.04-0.10 mm/s over this time period and ozone decreases of 0.15-0.35 ppm. SOCOL appears
to stand out from this group with significantly larger increases in upwelling and ozone decreases,
which act in concert to produce the very large temperature reductions seen in Figure 3e. The
impact of this larger change in upwelling in SOCOL can also clearly be seen in both total
column ozone change (see Figure 1b), and partial lower stratospheric ozone change (see Figure
1h). In general, differences in the change in upwelling explain differences in lower stratosphere
tropical ozone in the CCMVal-2 models. Also, this linear relationship exists when using the
same level for ozone and \( w^* \) (both at 50 and 70 hPa).

In the midlatitude lower stratosphere transport is also thought to play an important role
for ozone changes, which are generally positive over the 21\textsuperscript{st} century, however, the exact role is
not quantified in the present analysis (see further discussion below).

4. Polar ozone

We now consider changes in polar ozone. Austin \textit{et al.} [2010b] evaluate the CCMVal-2
model simulations of the Antarctic ozone hole in detail, and describe both the processes that are
well simulated and the areas that need additional work. More information on model biases,
especially those involving temperature thresholds for PSC formation, are provided in Austin et al. [2010b].

We will first focus on annual average changes and then look at a more detailed analysis of spring when the largest ozone depletion is observed. Figure 7 shows the evolution of annual average southern and northern polar ozone for total column ozone (upper panels, 7a,b), upper partial column (middle, 7c,d) and lower partial column (lower 7e,f). Again, we show ground-based total column ozone measurements (solid black curves, updated from Fioletov et al., [2002]) with respect to 1964 in Figures 7a,b.

Evolution of the upper partial columns is very similar to that of midlatitudes (see Figures 1d,f). However, the evolutions of the lower and total columns differ. All models show larger peak ozone loss around 2000 in the southern polar region than in the northern polar region. This is expected and seen in observations, as there is greater polar stratospheric cloud (PSC) formation in the southern polar region and a more stable polar vortex that lasts into spring each year. In general there are larger increases in ozone amounts by 2100 compared to 1960 in the northern polar region. It is difficult to say quantitatively with the existing simulations how much of the differences seen by 2100 in the total and partial lower stratospheric column ozone are due to polar chemistry differences, circulation differences, or a combination of the two. Additional simulations with fixed low chlorine concentrations or fixed GHG concentrations could help to quantify the relative impacts.

The largest ozone losses occur in the spring in the lower portion of the stratospheric column so we focus on this time and region in more detail. Figure 8a shows the Antarctic (60-90°S) lower stratospheric column ozone for October. Peak ozone loss averages about 100 DU around 2000 with a range from models between 70 and 170 DU. We applied a linear regression
analysis to the lower stratospheric column ozone amounts to calculate the contribution from
Cl<sub>y</sub>+αBr<sub>y</sub> using α=60, as appropriate for the polar lower stratosphere.

Figure 8b shows the contribution of Cl<sub>y</sub>+αBr<sub>y</sub> and the residual (Figure 8c), which is much
smaller and for most models typically less than ±20 DU. Cl<sub>y</sub>+αBr<sub>y</sub> dominates the partial column
ozone until the end of the 21<sup>st</sup> century when the residual is in most cases of comparable
magnitude. The decrease in mean age of air (i.e. stronger circulation, see Figure 8e) by itself and
in combination with upper stratospheric ozone returning to above historical levels acted to cause
increased ozone change (i.e, positive residual), however this could be tempered by the cooling of
the polar lower stratospheric temperatures (Figure 8f shows 50 hPa temperatures) which occurs
in nearly all models.

Figure 9a shows the Arctic (60-90°N) lower stratospheric column ozone for March. We
also note that this area is typically bigger than the size of the polar vortex so this average also
includes mixing effects (also to a lesser extent in the Southern Hemisphere during October).
Peak ozone loss averages at 30 DU around 2000 with a range between 10 and 60 DU. Again we
linearly regressed the Cl<sub>y</sub>+αBr<sub>y</sub> against the partial lower stratospheric column ozone. Figure 9b
shows the contribution of Cl<sub>y</sub>+αBr<sub>y</sub> and the residual (Figure 9c), which in this case is generally
larger with a consistent 30-40 DU residual by 2100 for most models. This seems to indicate that
in the NH polar regions circulation changes cause a larger increase in ozone than seen in the SH
by 2100.

5. Ozone Recovery
The above differences in ozone return dates between regions are summarized in Figure 10 which shows the evolution of the multi-model mean (MMM) total and partial column ozone, with respect to 1960 for high latitudes (solid curves) and midlatitudes (dashed curves). Total column ozone for the MMM (Figure 10a) shows a return to 1960 values by the 2030s in the NH in both mid and high latitudes. In the SH the total column ozone MMM exhibits a return to 1960 values by the 2060s in the midlatitudes and by 2080 at high latitudes. Nearly all the differences in total column ozone evolution are from the lower portion of the column (500-20 hPa, see Figure 10c) with very similar changes in both hemispheres occurring in the upper portion (Figure 10b).

Comparing with the evolution of halogens (red curves; using 60-90°S at 50 hPa as a proxy representative value), we see a significant difference in the dates ozone and halogens return to historical levels (i.e., 1960 levels). The MMM Cly+αBry does not return to 1960 values by 2100. This is later than the return of ozone (for extratropical regions). The cause of the earlier return of ozone in midlatitudes (shown in Figure 16 in Austin et al., 2010a) in both hemispheres is likely from an increased circulation [Austin and Wilson, 2006; Waugh et al., 2009; Li et al., 2009] along with increased upper stratospheric ozone from stratospheric cooling.

There is a noticeable difference in the dates when northern and southern hemisphere ozone returns to 1960 values (with no such difference in Cly+αBry, not shown). This interhemispheric difference is likely due to interhemispheric differences in transport. Some of the delay in the SH return is likely from asymmetries in polar ozone loss since there is significant polar ozone loss occurring in the SH in the early 21st century when NH ozone is returning to 1980 levels (not shown with reference to 1980 levels), causing less ozone to be transported into SH midlatitudes when the vortex breaks down [Atkinson et al., 1989]. The impact of polar ozone
losses being transported into the midlatitudes likely play the dominant role in the later return to historical values seen in the SH midlatitudes during first half of the 21\textsuperscript{st} century with any asymmetries in the strengthening of the stratospheric circulation likely having a larger impact toward the later half of the 21\textsuperscript{st} century.

6. Conclusions

Simulations of the past and future were performed using 14 chemistry-climate models for the CCMVal-2 activity. Austin et al. [2010a] have examined the evolution of total column ozone and compared the recovery of column ozone and Cl\textsubscript{y} in these models. Here we have contrasted the evolution in the upper and lower stratosphere, and examined the cause of the ozone changes (and differences among the models) particularly for the 21\textsuperscript{st} century.

The simulations presented here and in Austin et al. [2010a] show that there is some general agreement in the ozone evolution among the models, with all showing column ozone decreasing from 1960 to around 2000, and then increasing over the first half of the 21\textsuperscript{st} century. Models also show that the column ozone evolution varies with latitude, especially in the latter half of the 21\textsuperscript{st} century. There are clearly some quantitative differences in ozone evolution across the CCMVal-2 simulations, e.g., a large spread in simulated return of ozone to historical values [Austin et al., 2010a].

Separation into partial columns above and below 20 hPa reveals that the latitudinal differences in the evolution of the column ozone are almost completely due to differences in the lower stratosphere (the region of the atmosphere below 20 hPa). In all models, there are only weak latitudinal variations in the evolution of upper stratospheric ozone, and at all latitudes upper stratospheric ozone increases throughout the 21\textsuperscript{st} century and returns to 1960 levels before
the end of the century. There is, however, a large spread in dates of return to historical values, with, for example, dates of return to 1960 levels at 5 hPa varying from 2020s to 2060s. A multiple linear regression analysis indicates that the upper stratospheric ozone increase comes from almost equal contributions of decrease in halogens (Cl$_2$+αBr$_2$) and cooling from increased greenhouse gas concentrations (for the A1b greenhouse gas scenario considered in these simulations), with only small contributions from NO$_x$ and HO$_x$.

In the tropical lower stratosphere there is a steady decrease in ozone through the 21st century in all models, whereas ozone in middle and high latitudes increases during the 21st century. As a consequence tropical column ozone does not return to 1960 levels, whereas extratropical ozone returns or is close to 1960 levels before the end of the century. The decrease of tropical lower stratospheric ozone is due to an increase in tropical upwelling. Models with larger ozone decreases have larger increases in upwelling. Changes in transport also play an important role in the evolution of mid-latitude lower stratospheric ozone, and strongly contribute to the earlier return of ozone than Cl$_2$+αBr$_2$ to historical values.

There are quantitative differences between the hemispheres in the simulated ozone evolution. In particular, ozone returns to historical values earlier in the northern hemisphere. This difference in hemispheric return dates varies between 10 and 50 years depending on the latitude of interest and the reference year (e.g. 1960 or 1980) chosen, and is almost completely related to differences in the lower stratosphere. Additional simulations would need to be completed in order to quantitatively attribute causes for hemispheric differences. However, based on existing simulations it appears that the larger ozone loss from polar chemistry in the SH and the transport of this ozone depleted air into the midlatitudes as the vortex breaks down is one cause, especially
during the first half of the 21st century. Changes in the stratospheric circulation could also play a role in these differences.

At high latitudes, especially in the SH (Figure 8) Cl₂⁺αBr₂ dominates long-term trends in the lower portion of the column ozone (and total column ozone) with a small (generally positive) residual. This residual is likely caused by circulation changes and upper stratospheric return to above historical levels.

These results reinforce the conclusions in Eyring et al. [2007] using simulations from the CCMVal-1 activity: decreasing levels of halogens, continued stratospheric cooling, and changes in circulation are the major factors driving 21st century ozone trends. It is important to keep in mind that all of these simulations are based on a single GHG and halogen scenario, with only one model including an interactive ocean. In addition, the use of mixing ratio based halogen boundary conditions rather than emission-based constrains the models response [Douglass et al., 2008]. Future work should be done to explore these issues and their impact on ozone evolution in more detail.

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Table 1. Model description and references.

<table>
<thead>
<tr>
<th>Model</th>
<th>Atmospheric GCM</th>
<th>Domain/Resolution or Truncation</th>
<th>SST/Sea Ice for Ref-B2</th>
<th>Model Reference</th>
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<tr>
<td>AMTRAC3</td>
<td>AM3</td>
<td>Variable, ~200 km, 48 L, 0.017 hPa</td>
<td>CM2.1</td>
<td>Austin and Wilson (2010c)</td>
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<td>CAM3.5</td>
<td>CAM</td>
<td>1.9° x 2.5°, 26 L, 3.5 hPa</td>
<td>CCSM3</td>
<td>Lamarque et al. (2008)</td>
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<tr>
<td>CCSR-NIES</td>
<td>CCSR/NIES AGCM 5.4g</td>
<td>T42, 34 L, 0.012 hPa</td>
<td>MIROC / IPCC-AR4</td>
<td>Akiyoshi et al (2009)</td>
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<tr>
<td>CMAM</td>
<td>AGCM3</td>
<td>T31, 71 L, 0.00081 hPa</td>
<td>Interactive</td>
<td>Scinocca et al. (2008); deGrandpre et al. (2000)</td>
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<td>CNRM-ACM</td>
<td>ARPEGE-Climate version 4.6</td>
<td>T42, 60 L, 0.07 hPa</td>
<td>CNRM-CM3 AR4</td>
<td>Déqué (2007); Teyssèdre et al. (2007)</td>
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<tr>
<td>GEOSCCM</td>
<td>GEOS5</td>
<td>2° x 2.5°, 72 L, 0.015 hPa</td>
<td>HadISST1 for Ref-B1 and CCSM3 for Ref-B2</td>
<td>Pawson et al. (2008)</td>
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<tr>
<td>LMDZrepro</td>
<td>LMDZ</td>
<td>2.5° x 3.75°, 50 L, 0.07 hPa</td>
<td>OPA (ocean), LIM (ice)</td>
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<td>MRI</td>
<td>MJ98</td>
<td>T42, 68 L, 0.01 hPa</td>
<td>MRI-CGCM2.3.2</td>
<td>Shibata and Deushi (2008a,b)</td>
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<td>SOCOL</td>
<td>MAECHAM4</td>
<td>T30, 39 L, 0.01 hPa</td>
<td>ECHAM5-MPIOM</td>
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<td>ULAQ</td>
<td>ULAQ-GCM</td>
<td>R6/ 11.5° x 22.5°, 26 L, 0.04 hPa</td>
<td>CCM3</td>
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<td>UMSLIMCAT</td>
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<td>HadGEM1</td>
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<td>UMUKCA-METO</td>
<td>HadGEM-A</td>
<td>2.5° x 3.75°, 60 L, 84 km</td>
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<td>UMUKCA-UCAM</td>
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<td>WACCM</td>
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<td>Garcia et al. (2007)</td>
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Figure 1. Annual average total and partial column ozone amounts over 3 regions including the tropics (25°S-25°N) and midlatitudes of each hemisphere (35-60°S and °N). The partial column ozone is separated into an upper portion (d,e,f) from 20 to 0.1 hPa and a lower portion (g,h,i) from 500 to 20 hPa. All data are from 1960 to 2100 (except UMUKCA-METO to 2083) and have been smoothed with a 1:2:1 filter iteratively 30 times. Note the scale change in y-axis in panels d,e,f which are half the magnitude of the other panels. Ground-based total column ozone observations (black curves) are shown over 1964-2007 relative to 1964.
Figure 2. Evolution of ozone (a) in the tropics (25°S-25°N) at 5 hPa and change in ozone (b) with respect to 1960 levels for the CCMVal models. Models change in Cl$_y$+Br$_y$ (c) and temperature (e) with respect to 1960 levels. Contribution of changes in Cl$_y$+Br$_y$ (d) and temperature (f) to changes in ozone. All models are shown from 1960 to 2100 (except UMUKCA-METO to 2083) and have been smoothed with a 1:2:1 filter iteratively 30 times.
Figure 2 (cont.). Model evolution of NO\textsubscript{y} (g) and HO\textsubscript{x} (i) with respect to 1960 levels over (25°S-25°N) at 5 hPa. Contribution of changes in NO\textsubscript{y} (h) and HO\textsubscript{x} (j) to changes in ozone. All models are shown from 1960 to 2100 (except UMUKCA-METO to 2083) and have been smoothed with a 1:2:1 filter iteratively 30 times.
Figure 3. Profiles of ozone (a) in the tropics (25°S-25°N) for 2000 and differences in ozone (b) from 2000-2100 for the CCMVal models. Model differences in Cl\textsubscript{y}+5Br\textsubscript{y} (c), temperature (e), and NO\textsubscript{y} (g) from 2000 to 2100. Also shown are the contributions of changes in Cl\textsubscript{y}+5Br\textsubscript{y} (d), temperature (f), and NO\textsubscript{y} (h) to changes in ozone. Except UMUKCA-METO change shown from 2000 to 2083. Also shown in (a) are observations from MLS (solid black curve) for reference.
Figure 4. Tropical (25°S-25°N) profiles of the sensitivity of ozone to changes in (a) Cl\textsubscript{y}+5Br\textsubscript{y} (ppm/ppb), (b) temperature (ppm/K), and (c) NO\textsubscript{y} (ppm/ppb).
Figure 5. Comparison of zonal, monthly mean profiles of radicals from CCM models (coloured lines) versus 24-hour average radical profiles found using a PSS box model constrained by profiles of T, O₃, H₂O, CH₄, CO, NO₃, Cl, Br, and sulfate SAD from the various CCMs for the two indicated times and locations. The PSS model was run at CCM model levels from the tropopause (dashed colored lines) to 1 hPa. All simulations used JPL 2006 kinetics. The colored error bars represent the standard deviation about the zonal monthly mean for various days used to compute the mean. The black error bars represent the sensitivity of PSS output to variability in the CCM profiles of radical precursors. The metrics (“values of g”) were found as described in Chapter 6 of SPARC CCMVal2 [2010]. For the MRI model, PSS simulations are shown with and without loss of ClO by the reaction ClO+OH→HCl+O₂.
Figure 6. Scatter plot showing the change (from 1960-2100) in 70 hPa $w^*$ and 50 hPa ozone. The values are annual averages over 25°S-25°N for 12 of the CCMVal models with the black line showing the linear fit. UMUKCA-METO change shown from 1960-2083.
Figure 7. Annual average total and partial column ozone amounts over the high latitudes of each hemisphere (60-90°S and °N). The partial column ozone amounts are separated into an upper portion (c,d) from 20 to 0.1 hPa and a lower portion (e,f) from 500 to 20 hPa. All shown from 1960 to 2100 (except UMUKCA-METO to 2083) and have been smoothed with a 1:2:1 filter iteratively 30 times. Note the scale change in y-axis in c,d. Ground-based total column ozone observations (black curves) are shown from 1964-2007 with respect to 1964.
Figure 8. Evolution of ozone (a) for October partial column ozone (500-20 hPa) over 60°-90°S with respect to 1960 levels for the CCMVal models. Model contribution of Cl\textsubscript{2}+60Br\textsubscript{2} at 50 hPa (b) and residual (c) with respect to 1960 levels. Changes in Cl\textsubscript{2}+60Br\textsubscript{2} at 50 hPa (d), change in mean age of air (e) for models that included age of air tracer, and (e) change in temperature. All shown from 1960 to 2100 (except UMUKCA-METO to 2083) and have been smoothed with a 1:2:1 filter iteratively 30 times.
Figure 9. Evolution of ozone (a) for March partial column ozone (500-20 hPa) over 60°-90°N with respect to 1960 levels for the CCMVal models. Model contribution of Cl$_2$+60Br$_y$ at 50 hPa (b) and residual (c) with respect to 1960 levels. Changes in Cl$_2$+60Br$_y$ at 50 hPa (d), change in mean age of air (e) for models that included age of air tracer, and change in temperature (f). All shown from 1960 to 2100 (except UMUKCA-METO to 2083) and have been smoothed with a 1:2:1 filter iteratively 30 times.
Figure 10. Multi-model mean (MMM) of total (a) and partial column (b,c) ozone amounts for mid (35-60°S and 35-60°N, dashed curves) and high latitudes (60-90°S and 60-90°N, solid curves) and Cly+αBry for high latitudes (60-90°S) for reference.